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ANNUAL REVIEW (1959) OF LITERATURE ON RADIOACTIVE WASTES

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Robert A. Taft Sanitary Engineering Center
Cincinnati, Ohio

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ANNUAL REVIEW OF LITERATURE ON RADIOACTIVE WASTES

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PREPARED FOR THE RESEARCH COMMITTEE
OF THE
WATER POLLUTION CONTROL FEDERATION

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U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
PUBLIC HEALTH SERVICE
BUREAU OF STATE SERVICES
DIVISION OF RADIOLOGICAL HEALTH

ROBERT A. TAFT SANITARY ENGINEERING CENTER
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INDUSTRIAL WASTES - RADIOACTIVE

General

A prodigous amount of information on radioactive waste disposal and its effect on man's environment was made available last year. The United Nations released the proceedings on the Second United Nations International Conference on the Peaceful Uses of Atomic Energy which contained the papers presented at the Conference in Geneva in 1958. Of national import were the Hearings before the Special Subcommittee on Atomic Energy, Congress of the United States, 86th Congress, First Session on Industrial Radioactive Waste Disposal, January 28, 29, and 30 and February 2 and 3, 1959, which resulted in some 3,000 pages of testimony covering all phases of the atomic energy waste disposal program. Testimony of the second session of hearings covering the area of disposal of radioactive wastes into the ocean has not been released. In November, 1959, a conference was held on atomic energy waste disposal practices in Monaco. The proceedings of that conference are not yet available.

This review is organized under the subject headings: mining and milling operations; fuel preparation and fuel element manufacture; fuel reprocessing; waste handling, treatment, and disposal; costs; and legislation.

Mining and Milling Operations

Recovery of uranium from its ores in the United States is performed by hydrometallurgical processes. Liquid wastes may contain iron and manganese, sulfate, nitrate, magnesium, uranium, chloride, and dissolved solids as chemical pollutants and natural uranium, thorium-234, thorium-230, and radium-226, as radioactive pollutants (54). Severity of pollution depends on the size, quality, and ultimate usage of the receiving waters, while the legal aspects depend on the specific state laws involved. Holaday (53) lists the following compounds in uranium ores: free silica, vanadium, arsenic, or selenium, all of which are known to be toxic and are found in the wastes from the uranium mill. Wastes discharged per day amount to 12.24 mgd and contain 9.18 curies of radium.

Uranium, contained in effluents having a pH of at least 3, may be adsorbed at 80-90°C by passing over filtering beds made up of dibasic or tribasic alkaline earth phosphates (48). The tribasic phosphate of calcium and mixed alkaline earth phosphates (such as double phosphates of calcium and barium or their hydrolysis products) are particularly effective. A primary factor in retention is the pH of the effluent. Uranium can also be removed by agitation in the presence of a precipitate of a suitable adsorbing phosphate, and then the concentrate may be separated by filtration or settling.

Experiments carried out by Crespi, et al.(14), show that uranium contained in mother liquors and washings from precipitation of uranium peroxide may be recovered by concentration with ion exchange resins; uranium may then be precipitated from the concentrated solution as ammonium diuranate. Two particular advantages are referred to 1) simultaneous elution and chemical regeneration of the exchanger and 2) final separation of the uranium in the solid state combined with regeneration of the eluant.

The first of a series of studies planned by the Public Health Service to obtain data on characteristics of wastes, particularly radioactive, arising in uranium ore extraction, described operation of a resin-in-pulp mill (64). In this mill, the ore is crushed and ground, leached with sulfuric acid to dissolve the uranium, and the coarse sands separated from the slurry which is fed to ion-exchange resin columns. The uranium is extracted from the feed solution by resin beads, is stripped from the beads, precipitated, and dried to form yellow cake—the final product of the process. After stripping of uranium, the ion exchange feed solution becomes the slimes tails and is neutralized with lime before discharge to the slimes ponds. Data are presented on radium levels throughout the process. The authors estimate that 310 µg/day of dissolved radium went to the slimes pond, about 60 µg/day to the sands pond, and about 150 µg/day left the mill in the dried yellow cake.

In uranium ore milling operations, water consumption ranges from 0.2 to 1.4 with an average of about 0.6 gpm per ton of ore per day (29).

This amounts to 865 gallons of water per ton of ore treated and is equivalent to about 18 million gallons of liquid tailings per day from the industry.

Methods for the removal of radium from mill effluents were discussed including precipitation agents, ion exchange, and percolation through barite (18).

By means of neutralizing the slime tailings with lime, one mill (26) was able to precipitate about 85 per cent of the dissolved radium.

Fuel Preparation and Fuel Element Manufacture

Manufacturing operations in the fabrication of uranium or uranium alloy fuel elements may involve melting, casting, rolling, machining, and metal cleaning by the use of aids (35). Wastes generally are collected for the recovery of uranium. Residual wastes from metal cleaning, etching operations, and final rinsing solutions are monitored and diluted before release into the river.

At the gaseous diffusion plants at Oak Ridge and Paducah, the waste liquids discharged consist primarily of raffinate, spent chemical solutions, and very dilute aqueous solutions originating from cleaning equipment. Emlet (23) stated that the waste disposal operations at the Oak Ridge Gaseous Diffusion Plant were concerned exclusively with uranium. Alpha activity in the Clinch River one mile below the discharge point ranged from 2 to 5 x 10^{-9} µc/ml compared to a background alpha level of about 7 x 10^{-13} µc/ml. The wastes from the Y-12 area in Oak Ridge consist of wastes from spent process solutions--raffinates, supernatants, pickling and plating solutions, or from decontamination or cleaning operations, housekeeping procedures, etc. Creek alpha activity levels are less than 4.5 x 10^{-7} µc/ml after dilution. More concentrated wastes (alpha activity less than 7.2 x 10^{-2} µc/ml) are impounded in settling basins. Alpha activity levels in the creeks in the vicinity of Paducah ranged from approximately 10^{-7} to 10^{-8} µc/ml.

The feed materials plant at Fernald, Ohio, combines the various metallurgical processes essential to the production of reactor fuel elements from uranium ores and mill concentrates (32). The processes include: acid digestion of the ores and concentrates, organic phase extraction of uranyl nitrate, subsequent conversion of the nitrate to uranium oxides and tetrafluoride, reduction to uranium metal, and fabrication of the metal into fuel elements. The wastes produced contain varying amounts of naturally-occurring radionuclides, principally those occurring in the uranium series. Facilities provided (32) include hold tanks for raffinates, filters for removal of nitric insoluble material, an evaporator for concentration of the filtered raffinate, drum dryer, and a spray and rotary calciner for converting residual nitrates to metal oxides. Substantially complete removal of the radium is achieved by barium sulfate co-precipitation.

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Fuel Reprocessing

Because of the difficulty in dissolving some of the fuel cladding materials used with power reactor fuels (stainless steel, Zircaloy or zirconium, etc.), more study has been given to mechanical decladding in processing of spent fuels. Three approaches to mechanical processing are indicated.(3): sawing, shearing, and pulverizing. Selective chemical dissolution of fuel jackets and structural components is an attractive alternative to mechanical dejacketing or complete dissolution. Fuel cores thus exposed can be dissolved in another (probably less corrosive) reagent in a second step (3).

For Zircaloy-clad fuel elements having either an oxide core or a uranium-niobium-zirconium alloy core, the Zirflex process (6 M ammonium fluoride - 1 M ammonium nitrate dissolving agents) is under investigation (3). The Sulfex (sulfuric acid) process is under test for processing stainless steel clad fuels. At the Savannah River Plant, aluminum jackets of various shapes are separated from uranium by a boiling aqueous solution of sodium hydroxide and sodium nitrate.

Simultaneous dissolution of jacket and core in the case of zirconium-containing fuels can be accomplished by hydrochlorination (Zircex process), by use of aqueous fluoride-nitric acid mixtures, or by dissolution in hydrofluoric acid-hydrogen peroxide. Stainless steel fuel elements are dissolved in nitric acid-ammonium bifluoride mixtures (Niflex), in dilute aqua regia (Darex), and in dilute sulfuric acid (Sulfex). Mercuric nitric acid is under study for dissolution of aluminum fuel elements.

The Redox and Purex processes use methyl isobutyl ketone and tributyl phosphate, respectively, as extractants and are used in the recovery of uranium and plutonium from production reactor fuels. Work continues to improve these processes and to adapt them to the processing of nonproduction reactor fuels (3). Redox is being considered for Darex feeds (stainless-steel fuels dissolved in 5 M nitric acid - 2 M hydrochloric acid), but the results have not been entirely satisfactory. Niflex feeds subjected to Redox processing were also poorly decontaminated.

A principal limitation of Purex decontamination is ruthenium activity retained in product streams (3).

Ion exchange processes have been applied to the recovery of plutonium and neptunium, uranium, americium, and actinide elements and to the removal of radioactive zirconium and ruthenium from process solutions (3).

Several sites are working on pyrometallurgical processes in anticipation that fuel processing costs will be reduced. This anticipation arises from elimination of chemical conversions and the ability to handle short-cooled fuels. Methods for decontamination of thorium-uranium fuels include (3): 1) direct volatilization of many fission products in an arc-melting furnace using a nonconsumable tungsten electrode for removal of strontium, cesium, and appreciable quantities of the rare earths; 2) drip melting in which a thorium slug is suspended

in vacuum in a high frequency induction coil; 3) fused salt electrolysis; and 4) separation by magnesium extraction.

Homogeneous Reactor Processing

Reactors utilizing homogeneous fuel have the potential advantage that a side stream of fuel can be continuously removed and processed. Since such processing could conceivably be accomplished in a closed loop, modest decontamination is adequate. This concept is utilized in the aqueous homogeneous reactor at Oak Ridge (3).

Waste Handling, Treatment, and Disposal

General

Wolman (67) reported that in 1958 the atomic energy industry had invested somewhat over $23 million in treatment and release facilities for low level wastes. During the year, approximately 1.5×10^9 gallons of low level waste were discharged. The total curies of activity discharged to date have totalled somewhat over 2.6 million curies (approximately 95 per cent of total activity has been associated with Hanford Operations). The treatment and disposal of fuel processing wastes (high level wastes) continue to pose a difficult, expensive problem (67). Approximately 65 million gallons of high level wastes have been placed in storage and the investment in tank storage amounts to over $115 million.

Thomas (62) draws the following conclusions from studies made on Cochichewick Brook near Lawrence, Massachusetts, and on other stream systems: 1) Under favorable conditions substantial quantities of low level wastes may be released to inland waterways, 2) dispersion and so-called self-purification processes do not destroy radionuclides, but dilution to safe levels or transfer to other elements of the environment takes place, 3) uptake is a function of stable element concentration, 4) self-purification processes may make certain nuclides more amenable to treatment at water works, 5) storage of long-lived nuclides on stream bed is dangerous, and 6) hydraulic mixing and dispersion may be quite different during different periods of the year.

Using Columbia River radioactivity data and a mathematical model, Harris (30) estimated downstream radioactivity levels on the basis of decay. Selective uptake and deposition could be detected by comparing initial average decay rates observed at downstream stations with average decay rates predicted for these stations from a model fitted to the outfall sample.

The uptake of specific nuclides by aquatic communities was discussed by several authors (15,25,52). In discussing Hanford operations, Foster (25) stresses that hydrologic data alone is insufficient for accurate evaluation of radiological problems, but that a complete evaluation calls for detailed knowledge of the environmental, biological, and operational variables, and accurate nuclide analyses.

Studies on the Columbia River showed that significant quantities of various isotopes were concentrated in the bodies of most organisms which had access to the contaminants via the natural food web. Food organisms of man, therefore, could accumulate hazardous levels of certain radionuclides from water which contained concentrations of the contaminants that were well within the permissible limits for drinking water. The data presented (15) emphasize the value of radioecological research to further elucidate and evaluate environmental relationships pertaining to radioactive waste disposal.

Foster (24) states that in the evaluation of exposure, which man receives from an environment that contains radioactive waste,

consideration must be given not only to drinking water but to all sources which may contribute to the total exposure. For exposure from internal emitters, it is the burden of isotopes deposited in the body which is of importance. The MPC's are only guides to be used as one facet of exposure calculation. Because plants and animals concentrate certain isotopes, these forms must be included in the monitoring program since their contribution to the total exposure may exceed the contributions from other sources.

Studies regarding the identification, source, and control of radionuclides in the recirculating type, cooling water system of the Low Intensity Test Reactor (LITR) at the Oak Ridge National Laboratory were reported by Moeller, Leddicotte, and Reynolds (42).

In discussing the problem of waste disposal from nuclear powered vessels, Smith (57) indicates that only limited facilities must be provided for essentially complete protection of the ship's marine environment. The wastes expected from water-type reactor systems are such that intelligent use of the sea for direct disposal is possible.

Low level wastes are treated by a process including $Fe_2(SO_4)_3$ and NaOH addition, settling, filtration through quartz (sand), and ion exchange treatment through cation and anion exchange beds in series (65). The treated effluent is discharged to the river and the sludge goes to covered sludge storage beds.

A description is also given of a pilot plant designed to treat radioactive wastes from chemical, physical, and biophysical laboratories (8).

Describing experiences at Mol, Belgium, Dejonghe, et al. (16), indicate that "cool" effluents are treated by phosphate coagulation and that biological treatments can also be applied. Batch tests give decontamination results of more than 95 per cent for uranium and 80 to 95 per cent for most of the β-γ emitters. The "warm" effluents containing cationic radioisotopes can be easily and cheaply treated by filtration through columns or continuous filters of lignite (brown coal) after phosphate coagulation. If levels of activity are high, evaporation is the only practical method of treatment.

Following a discussion of the waste disposal practices -- both actual and speculative -- Struxness and Blomeke (58) conclude that the treatment of high level radioactive wastes and disposal of them in a permanent sense are mainly in the conceptual and formative stage. There has been little treatment given to high level wastes other than neutralization, evaporation, and storage as concentrated salt solutions in steel tanks.

Since solids can be stored with less risk of dissipation than liquids, the final aim according to Burns and Glueckauf (12) must be to reduce all wastes to two final components: 1) adequately pure water, and 2) solid radioactive waste. The most effective but also most

expensive way of carrying out this separation is by evaporation. For large volume-low activity wastes, some form of chemical treatment is more economical than evaporation. After single stage precipitation treatment, most of the cesium and iodine, some strontium, and some ruthenium usually remain. Further stages of treatment are, therefore, required. These can include further precipitations, preferably combined with treatments intended to convert anionic and non-ionic ruthenium species into cationic precipitable ones. Other types of second-stage treatment which can be carried out cheaply are: 1) passage through ion exchange beds specific for strontium or cesium, or 2) complete deionization. Three schemes were studied and costs estimated per 100,000 gallons (Imp.) as follows: 1) precipitation and controlled disposal (99.65 per cent of α and 90.89 per cent of β activity removed) - £123.25; 2) precipitation and vermiculite columns (99.95 per cent α and 99.36 per cent β activity removed) - £117; and 3) precipitation and electro-deionization (100 per cent α and 99.85 to 100 per cent β activity removed) - £144.1.

Continuous processing of the supernatant from neutralized sodium nitrate or aluminum bearing waste by ion exchange appears particularly promising for the removal of cesium and strontium, which have been removed by factors greater than 10^5 and 10^3, respectively (5). Since about 99 per cent of the strontium remains in the neutralized waste sludge, the overall removal for strontium was greater than 10^5. Similar results are expected for the other limiting long-lived isotopes,

$Pu^{239,240}$, Cf^{242}, and Am^{241}, based on their known solubilities in alkaline solutions and ion exchange characteristics. Ruthenium-106 can be removed by electro-deposition by factors greater than 200.

Frequently, statements have been made on the advantages to be gained in recovering long-lived fission products and then of releasing the residual wastes after short term detention -- up to 10 years or so. Moore and Burns (43) bring this question back into perspective in pointing out that most recovery processes will remove strontium and cesium with a decontamination factor of 10 to 100 whereas decontamination factors of 10^6 and 10^9 are needed to reduce the cesium and strontium to permissible drinking water levels. With decontamination factors of 10 to 100, complete confinement of the wastes, further treatment to reduce these isotopes to nonhazardous levels, or controlled disposal to the ground under favorable conditions of soil retention and remoteness from public waters would be required. Fission product utilization, even if highly successful, will have a relatively small effect on the waste disposal problem (40).

In commenting on the future of high level waste management from the nuclear power industry (to the year 2000 A.D.), Parker (51) stated that 1) the expanded use of atomic energy need not be inhibited at all by the radioactive waste disposal problem unless there should be concurrently a drastic reduction in permissible radiation exposures; 2) feasible methods at reasonable costs are known for improving control of wastes; 3) continuous examination of the exposure pathways around

each principal release site is needed; 4) extensive research into three areas of radiological sciences will be needed, namely: a) the mechanisms of transmittal of radioactive materials through numerous environmental pathways; b) the significant radiation exposure to man and other life forms as receptors from these channels; and c) acceptable limits of exposure of man and other life forms as individual receptors and as components of a genetic pool.

Wolman (68), in general summary, makes the following statement: The ". . . point with respect to low activity waste is simply that it is not a disappearing problem. It is a mounting one. Its greatest value is that it can be managed with economy, but it must be managed with rather careful criteria and extremely careful public supervision." Commenting further he goes on to say, ". . . I have been a strong advocate, in the case of other pollutants more familiar to us, of using nature, under control, as one of our major methods of disposal. I would establish and use exactly the same thesis with low level activity in the radiation field. I think we would be discarding an extremely valuable natural asset if we did not make use of nature with low activity waste. My only comment was that, when we do that, as we do with any other kind of pollutant, we must do it with logic, with criteria, and with persistent monitoring. That was not intended to indicate that we should stop doing it, because I think we would throw away an extremely valuable disposal method."

By example Swope (60) reviewed AEC regulations regarding low level wastes and described procedures used by Argonne in handling its waste. She makes a point which bears repeating, namely, " . . . to awake city, county, and state agencies to the fact that, if they have not already done so, they must be prepared to analyze for radioactivity."

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Tank Storage

Doan (19) describes the disposal facilities at the various reactor sites located at the National Reactor Test Station. Radioactive liquid wastes result from the three cycles of liquid extraction by which the enriched uranium is recovered from the dissolved fuel elements at the Idaho Chemical Processing Plant. After evaporation the wastes are stored in appropriate underground tanks -- water-cooled tanks for first-cycle wastes and uncooled tanks for second- and third-cycle wastes. Tank storage costs are given as $2.63 per gallon for stainless steel tanks in which acid wastes are stored and $4.90 per gallon for storage of zirconium wastes. At both the Materials Test Reactor and the Engineering Test Reactor sites (19), the bulk of the waste results from the primary process water system used to cool the reactor core components.

Chemical Treatment

In earlier studies of soil disposal at Hanford, it was observed that the retention of strontium-90 on certain soils was greatly enhanced in the presence of phosphate ion, while cesium-137 retention was unaffected. Further study showed that the mineral calcite ($CaCO_3$) was responsible for the increased strontium-90 retention (2). The mechanism involved was determined, and its applicability to strontium-90 removal from wastes was partially evaluated. It was found that the alkaline-earth carbonates were altered to carbonate-apatite in the presence of alkaline phosphate solutions. The mechanism shows promise in removal

of so-called "bone-seeking" radioisotopes from wastes, since this mechanism and bone formation both result in a carbonate-apatite containing these radioisotopes.

The process waste treatment plant at Oak Ridge National Laboratory was described by Cowser, Morton, and Witkowski (13). They indicated that the lime-soda softening process is an effective and economical method for removal of strontium and the total rare earths from large volumes of low level wastes. Cesium can be removed from these wastes by the addition of clay in this process. Results reported indicate 80 to 90 per cent removal of strontium; other nuclides showed removals of 50 to 90 per cent or more.

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Ion Exchange

Šušić and Selenić (59) used ion exchange techniques with different concentrations of HCl to separate long-lived radioisotopes from fission products and corrosion elements. Purification of the radioisotopes from the group is accomplished with Complexone-II for cesium-137,

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** See Op. Cit.

ammonium citrate for strontium-90, and anion-exchange resin, Dowex-2, with concentrated HCl for the isolation of zirconium-95-niobium-95. Uranium and iron, in the form of thiocyanate complexes, can be precipitated from a mixture containing fission products and corrosion elements by coprecipitation with methyl violet.

Basic silica gel (called a cheap ion exchanger) is capable of adsorbing strontium-90 and cesium-137 from wastes (41). Removal of cesium-137 may be increased to 99.5 per cent by passing the solution through a layer of silica gel.

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Fixation

The equipment designed and developed for sintering experiments at Oak Ridge National Laboratory was described by Boegly and Struxness (6).

Canadian experiments with nephaline syenite (66) showed that volatilization of ruthenium and cesium occurred. By preparing an

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** See Op. cit.

alkaline mix the nitrosyl nitrate complexes in the mix are hydrolized to a nitrosyl hydroxide of ruthenium which decomposes to the non-volatile ruthenium dioxide at temperatures below 900°C, thus retaining 99 per cent of the ruthenium. At temperatures above 900°C, ruthenium dioxide is air oxidized to the volatile tetroxide. Firing the mix in an atmosphere of CO_2 reduces the loss of ruthenium. Cesium losses as a function of temperature and mix composition are not as well understood. Equipment has been constructed to incorporate several thousand curies of aged fission products into glass. Batchwise treatment on a scale of about 100 curies is possible. Even though leach rates are low, leaching over a considerable period of time will result in the release of large amounts of activity. The suggestion is made of utilizing glazes to reduce leach rates or that the glass be stored in dry areas. From studies with glazes and fused ceramics, Eliassen and Goldman (22) conclude that aluminum-bearing fission product wastes can be incorporated into ceramic glazes, thus getting rid of the aluminum nitrate from chemical processing as well as the fission products. The systems showing greatest promise to date are: $CaO-Al_2O_3-B_2O_3-SiO_2$ and $Na_2O-CaO-Al_2O_3-B_2O_3-SiO_2$.

The French (9) have reported results of studies on the retention of activity in glasses prepared at 1000°C and micas at 1300°C.

Thomas (63) examined the stability of radioactive glasses and sinters theoretically and the problem relating to the mobility of radioactivity within masses of earth.

A process utilizing clay sponges is described by the Los Alamos Scientific Laboratory (39). The process involves the preparation of a highly porous clay body, which is fired at about 800°C, soaked in the liquid waste, dried, resoaked (the cycle is repeated as many as 3 to 5 times), and finally fired at about 1300°C to permanently fix the radionuclide. Studies show that the sponge has absorbed a Purex waste up to 200 per cent of its weight after four cycles.

Brookhaven National Laboratory reports (31) that the efficiency of clay columns for high level waste decontamination depends on the concentration of nonradioactive cations in the influent solutions. Waste solutions are concentrated, deacidified, and when treated with thioacetamide should be compatible with the clay process and should yield high decontamination factors for strontium and cesium (31).

Kaufman and Garden (37) add Portland cement and vermiculite to absorb liquid wastes. The solidified waste is stored and handled as a solid waste package.

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Calcination

Calcination is designed to meet two basic objectives of ultimate disposal: immobilization and minimum volume (20). In the calcination process the salts present in the wastes are converted to their stable oxides by thermal decomposition and steam hydrolysis. The three stages of the process include: 1) calcination in a rotary ball kiln; 2) off gas recovery; and 3) oxide leaching and storage. The authors conclude that calcination in rotary ball kilns appears to be practical with respect to product quality, mechanical design, off gas volume, and dust carryover.

Continued developments in the fluidized bed process of fuel processing is described by Loeding, et al. (38). Comparative economic studies have been made and show that fluid bed calcining will cost $0.582/gallon and ball mill calcining, $0.863/gallon.

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* See Op. cit.

At Argonne, feasibility studies are being performed on the fluidized-bed calcination of Hanford Purex low acid waste solution (3).

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Ground Disposal

In commenting on ground disposal, Theis (61) stated that the safe and efficient disposition of waste demands: 1) a thorough exploration of the geology and hydrology at the site; 2) an extension of our basic knowledge of movement of ground water and the geochemistry involved; and 3) a follow-up monitoring program to insure that the hydrology and geology have been properly interpreted.

Disposal of low-level liquid radioactive wastes to the ground at Hanford and the Savannah River Plant is founded on the ability of various earth materials to remove and retain fission product cations. Practical experience covering 12 years at Hanford and four years at Savannah is reviewed (11). Oak Ridge practices in ground disposal are described by de Laguna, Cowser, and Parker (17).

** See Op. cit.

At the National Reactor Test Station, low level wastes are discharged directly through disposal wells which penetrate the ground water table, or indirectly through seepage pits or ponds (34). It is reported (34) that during the period 1952 through 1958, 8,900 curies of radioactivity were released to the ground of which 85 per cent had a half-life of less than 10 days; 10 per cent, 10 to 100 days; and 5 per cent, more than 100 days.

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Deep Well Disposal

Deep well disposal of radioactive wastes is considered feasible and economical for low and intermediate level wastes (6). In selecting a site for injection, the geology and hydrology must be known and monitored subsequently. Chemical treatment of wastes will be necessary prior to injection.

The Subcommittee on Disposal of Radioactive Waste, American Petroleum Institute considered the problems associated with the disposal of radioactive waste in deep wells (1). Among the principal problems to be solved were: 1) the disposal reservoir must be clearly defined and capable of confining the radioactive waste solution for indefinite periods; 2) the heat generated during decay should be dissipated at a sufficient rate to prevent excessive increases in temperature; 3) the disposal formation should be protected against damage such as fracturing, premature plugging, and other causes; 4) the failure of disposal well equipment, as a result of corrosion or other causes, must be prevented; and 5) an economical disposal plant must be designed and safe operating procedure must be developed.

Salt Disposal

Consideration of the use of salt cavities for storage of radioactive wastes resulted from the knowledge and experience of storage of petroleum gases at such sites since 1950 (1). Salt formations have the following advantages when considered for storage of radioactive wastes:

1) impermeability; 2) wide distribution of areas; 3) large volumes of salt close to the surface; 4) acceptable structural properties; 5) low thermal conductivity; 6) wastes can be contained; 7) low cost of developing space; and 8) location in areas of low seismicity. Offsetting these advantages is the fact that little is known about the degree to which the desired structural properties of the salt are affected by liquid wastes, pressure, temperature, and radiation.

Studies of thermal consideration by Birch (4) indicate that nearly any quantity of waste producing heat at the rate of 0.01 watts per gallon, if distributed in a layer of the order of 100 meters thick, can be accommodated without undue rise of temperature. Schechter and Gloyna (56) also have reported on theoretical studies of temperature distribution in salt cavities. They conclude that cavities much in excess of 30 feet in radius will produce temperature increases in waste beyond permissible limits, that the wastes should be diluted to reduce temperature rise, and that the temperatures are sensitive to changes in values of vapor pressure, total volume occupied by the waste, and thermal conductivity.

Gloyna, et al. (28), summarized research on the storage of reactor fuel wastes in salt formations. A few of the problems investigated include: 1) geological studies of salt domes; 2) structural stability; 3) permeability; 4) thermal heat effect; 5) creep; 6) stress distribution; 7) time effect; 8) chemical effect; 9) radiation effect; 10) cost of developing solution cavities; and 11) development of design principles for disposal into salt cavities.

Analytical studies (49) show that it is possible to store two-year-old waste in a 10-foot diameter sphere without exceeding a temperature of 200°F. Laboratory tests show that the structural properties of rock salt are not greatly altered by high radiation doses although the creep rate is increased at high temperatures. Chemical interaction of liquid wastes with salt produces chlorine and chlorine compound gases, but the volume is not excessive. Field tests are planned to study migration of nuclides through the salt and the deformation of the salt cavity.

Ocean Disposal

Two reports relating to waste disposal into the sea were released by the National Academy of Sciences - National Research Council; one covers the possible disposal of radioactive wastes into selected areas of the Atlantic and Gulf Coastal waters (45), and the other deals with release of wastes from nuclear-powered ships (44).

Twenty-eight possible sites were recommended for offshore waste disposal of packaged, low-level radioactive wastes (45). For safe disposal the following criteria were indicated: survey of water circulation and inventory of bottom-living organisms; limitation of deposition to 250 curies per year and 100 curies per month of strontium-90 or its equivalent per site; separation of adjacent areas by at least 75 miles and limitation of three disposal areas per 300 miles of coast line; use of sinkable containers; and periodic monitoring.

Disposal of nuclear powered vessel wastes were considered on the basis of release to four marine environments: 1) harbors, estuaries, and coastal waters out to two miles from shore; 2) coastal area between two and twelve miles from shore; 3) outer Continental Shelf, extending 12 miles offshore outward to the 200-fathom-depth contour; and 4) open sea, beyond 12-mile limit and depth greater than 200 fathoms (44). Specific recommendations are presented to cover disposal operations in each of these areas.

Revelle and Schaefer (55) review the problems associated with sea disposal of radioactive wastes under the following headings: pollution hazards in confined waters through accidents to nuclear-powered ships; fate of radioactive materials in coastal waters; use of the deep ocean waters as possible sites for high level wastes; removal of radioactive materials from ocean waters by sedimentation processes; uptake and retention of radioisotopes by organisms used as food for man; and effects of the biosphere on the distribution and circulation of radioactive elements.

An area suggested for disposal of high level wastes has been deep ocean trenches. Bogorov and Kreps (7) studied the Tonga Trench, depth 10,816 m, and reported that there was mixing in the trench. Radioactive waste compounds dumped into the trench and dissolved could be inevitably brought to the upper layers of the ocean exploited by man. They do not recommend use of this or other trenches until extensive further oceanographic investigations are made.

The investigations carried out by the U. K. Atomic Energy Authority Establishment at Winfrith Heath to Study the possibilities of utilizing the area of the coast for low level waste disposal are described by Bowles, et al. (10).

Dunster (21) reviewed the first five years' experience in the release of low level radioactivity into the Irish Sea at Windscale during which discharges at a mean rate of 3000 curies per month took place. As a result of the experimental program, he feels that it would be safe to release some 20,000 curies/month of fission products at a point 3 km off this part of the Cumberland Coast. More recent data suggests that this could possible be increased to as much as 100,000 curies/month.

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Costs

Kaufman and Garden (36) have reported the following costs based on their 1957 operations: dry waste per drum - $6/ft.^3$; concrete block - $9.50/ft.^3$; and liquid waste per drum - $2/gal.

Comparative costs for sea and land disposal were made by Nielsen (17) for handling wastes from the Livermore Radiation Laboratory. Depending on the number of drums shipped, mode of shipment, and depth of disposal, costs of sea disposal ranged from $5.28 to $20.47 per 55-gallon drum. Land disposal costs varied from $14.05 to $19.25 per drum depending on place of disposal and mode of transportation used.

Because of the long period of time involved, present worth concepts (the time value of money) were used in economic calculations involving waste disposal (27). In comparing the three systems, 1) tank storage, 2) salt cavity storage, and 3) combined tank-salt cavity system, they showed that either a combination tank-salt cavity system or a cavity system alone leads to lower costs for liquid-waste storage than tank storage alone (neglecting transportation charges).

Legislation

The National Committee on Radiation Protection released a new handbook (Handbook 69) with revised recommendations for maximum permissible body burdens and maximum permissible concentrations of radionuclides in air and in water for occupational exposure (46).

In 1956, the Ministry of Housing and Local Government (England) invited the Radioactive Substances Advisory Committee (appointed under the Radioactive Substances Act, 1948) to appoint an expert panel with the following terms of reference: "To ascertain the nature and quantity of radioactive waste likely to arise in the forseeable future; to advise on the best methods of securing that the waste is disposed of safely; and to advise whether any new legal provisions or amendments to existing legislation are necessary to ensure safe disposal, and, if so, to advise on the form which the new provisions or amendments should take." Those principles which are of most interest to the Federation are quoted from the report (53).

"(i) It is not in the nature of radioactivity that the economic value of the useful materials involved can be realized without some release of ionizing radiations to the general environment. But this release can be controlled within safe limits.

"(ii) Discharges of waste should be so controlled as to ensure, irrespective of cost, that they will not put into circulation an amount of radioactivity which would directly endanger the health of any member

of the public living in the neighborhood. In addition in considering whether a given discharge should or should not be permitted, it is necessary to decide whether the radiation from the discharge when added to that from all other sources of radiation, is likely to result in a genetic hazard to the nation as a whole; from this point of view it is as much the concern of, for example, London as of Manchester if the inhabitants of either are exposed to ionizing radiations.

"(iii) In considering the permissible general levels of discharge, it is necessary to have regard to other additions to natural background activity - e.g. from X-rays or fallout - which are incapable of being accurately assessed or influenced except on a national basis.

"(iv) The numbers of people who have sufficient scientific knowledge to evaluate these problems is so limited that it would not be practicable to set up a large number of separate local controls.

"(v) The indications are, therefore, that control ought to be organized on a national basis as a government responsibility.

"(vi) This does not mean that the local disposal of wastes is undesirable. On the contrary, it is desirable that conventional methods should be used where practicable, since the object of harmless disposal to the environment can most often be met, and the increased exposure caused by multiple handling avoided, by such methods. The authority responsible for control should, however, have in reserve means of disposing of wastes which are too active to permit of safe local disposal."

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